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(54) **Cycloolefin type random copolymer compositions and uses thereof**

Ungeordnete Cycloolefincopolymer-Zusammensetzungen und deren Anwendung

Compositions de copolymères statistiques de cyclooléphines et leur application

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(56) References cited:  
**EP-A- 0 156 464 EP-A- 0 203 799**

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## Description

This invention relates to cycloolefin type random copolymer compositions which have outstanding characteristics such as transparency, heat resistance, moisture resistance and chemical resistance, and which have excellent high precision moldability as well as adhesion of information recording films.

Examples of known synthetic resins having excellent transparency are polycarbonates, polymethyl methacrylate and polyethylene terephthalate. For instance, polycarbonate resins have excellent transparency as well as excellent heat resistance, heat ageing characteristics and impact strength. However, polycarbonates have poor chemical resistance as they are easily attacked by a strong alkali. Polymethyl methacrylate is readily attacked by, for example, ethyl acetate, acetone, or toluene, swells in ether and, moreover, has low heat resistance. Though polyethylene terephthalate has excellent heat resistance and mechanical properties, it has a weak resistance to strong acid or alkali and is subject to hydrolysis.

Polyolefins which are widely used as general-purpose resins have excellent chemical resistance and solvent resistance as well as excellent mechanical properties. However, many polyolefins have poor heat resistance and poor transparency because they are crystalline resins. In general, to improve the transparency of polyolefins, nucleating agents are incorporated into polyolefins to render their crystal structure microcrystalline, or the polyolefins are quenched to stop the crystal growth. However, it is hard to say that the alleged effects obtained by these procedures are sufficient. Incorporating into polyolefins a third component such as nucleating agents involves the risk of marring the various excellent properties inherent in polyolefins, and the quenching procedure requires a large-scale apparatus and, in addition, involves the risk of the lowering heat resistance and rigidity as the crystallinity index of polyolefins decreases. That is, in either case, it is not possible to control perfectly the crystal growth of the polyolefins, and there remain problems, such as the molding shrinkage factor of the thus treated polyolefins is high, and the post-shrinkage of molded articles obtained after molding said polyolefins is also high.

Under such circumstances, a copolymer of ethylene and 2,3-dihydroxydicyclopentadiene has been disclosed as an example of copolymers of ethylene and bulky comonomers, for example, in U.S. Patent 2,883,372. However, this copolymer has poor heat resistance as it has a glass transition temperature in the vicinity of 100°C, though the polymer is well balanced between rigidity and transparency. A similar drawback is also observed in copolymers of ethylene and 5-ethylidene-2-norbornene.

Japanese Patent Publ. No. 14910/1970 proposes a homopolymer of 1,4,5,8-dimethano-1,2,3,4,4a,5,8,8a-octahydronaphthalene. The proposed polymer, however, has poor heat resistance and heat ageing characteristics. Japanese Patent L-O-P Publ. No. 127728/1983 further proposes a homopolymer of 1,4,5,8-dimethano-1,2,3,4,4a,5,8,8a-octahydronaphthalene or copolymers of said cycloolefin and norbornene type comonomers, which are apparently those obtained by ring opening polymerization (ring opening polymers) in light of the disclosure in said publication. These ring opening polymers which have unsaturated bonds in the polymer main chain, however, have poor heat resistance and heat ageing characteristics.

We have found that cycloolefin type random copolymers of ethylene and specific bulky cycloolefins are synthetic resins which are well balanced between heat resistance, heat ageing characteristics, chemical resistance, solvent resistance, dielectric characteristics and mechanical properties, and that said cycloolefin type random copolymers have an excellent performance in the field of optical materials such as optical memory discs and optical fibers. On the basis of the above findings, we have already made various technical proposals as disclosed in Japanese Patent L-O-P Publ. No. 168708/1985, Japanese Patent Appl. Nos. 220550/1974, 236828/1984, 236829/1984 and 242336/1984. In spite of their being olefin type polymers, the cycloolefin type random copolymers proposed have excellent adhesion to various materials. However, when these cycloolefin type random copolymers are used for molding information recording base boards under severe conditions, insufficient adhesion to information recording films has sometimes been found.

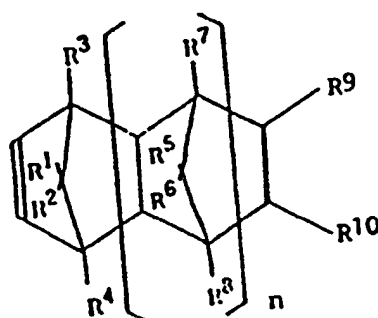
Information recording base boards (hereinafter sometimes abbreviated to "optical discs") are constituted from, for example, polymethyl methacrylates, polycarbonates, polystyrenes, rigid polyvinyl chlorides or epoxy resins. However, optical discs molded from these resin materials individually have a fairly large number of drawbacks. For instance, polymethyl methacrylates have poor heat resistance and high water absorption, as high as 0.4%. Therefore, molded articles obtained from polymethyl methacrylates are subject to dimensional change owing to their moisture absorption and are liable to warp. Polycarbonates have a high modulus of photoelasticity as well as double refraction, even though they have excellent in heat resistance. Moreover, polycarbonates have low surface hardness and are liable to marring, and the problem of moisture resistance still remains unsolved, though the water absorption of polycarbonates is as low as 0.15% in comparison with polymethyl methacrylates. Polycarbonates are practically unusable as optical discs, because they have poor heat resistance, impact resistance and surface hardness, not to speak of their high double refraction after molding. Polyvinyl chlorides are difficult to use as optical discs, because they have very poor heat resistance and, moreover, poor processability and service durability. Epoxy resins involve a difficulty in mass production as they have poor moldability, though they have excellent heat resistance. Moreover, epoxy resins have a high modulus

of photoelasticity and a problem of double refraction due to residual stress produced at the time when they are molded. Glass is fragile and easily broken and, moreover, has poor handling and productivity characteristics as it is relatively heavy, though it has excellent heat resistance, moisture resistance and surface hardness.

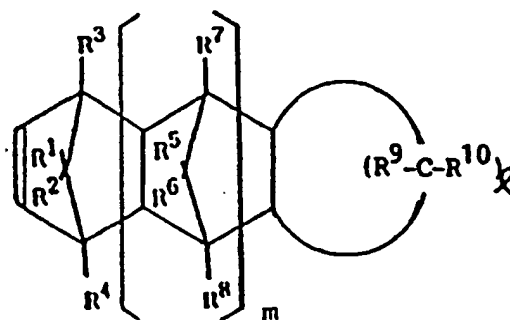
The present invention seeks to solve the problems associated with the prior art as mentioned above, and to provide resin compositions which have excellent transparency, heat resistance, heat ageing characteristics, chemical resistance, solvent resistance, dielectric characteristics, various mechanical characteristics and precision moldability and, moreover, which have excellent adhesion of information recording films.

The present invention provides a composition comprising

(A) a random copolymer containing units derived from ethylene and units derived from a cycloolefin of formula [I] or [II],



[I]



[II]

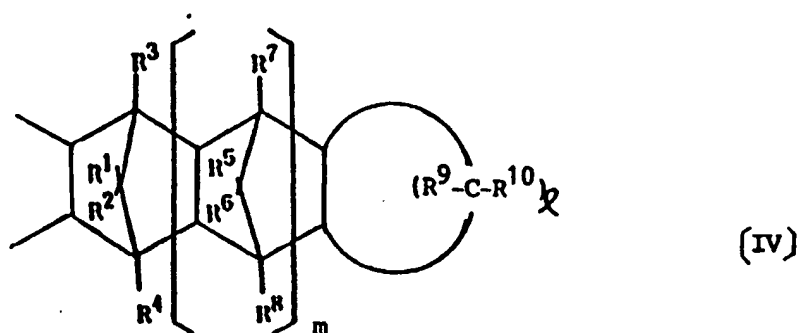
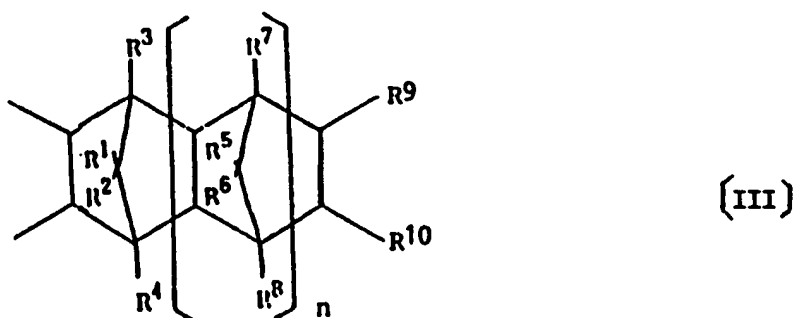
wherein  $n$  and  $m$  are each 0 or a positive integer,  $\ell$  is an integer of at least 3, and  $R^1$  to  $R^{10}$  each represent hydrogen, halogen or a hydrocarbon group, said random copolymer having an intrinsic viscosity  $[\eta]$  of 0.05-10 dl/g as measured at 135°C in decalin and a softening temperature (TMA) not lower than 70°C, and

(B) a random copolymer containing units derived from ethylene and units derived from a cycloolefin of said formula [I] or [II] and having an intrinsic viscosity  $[\eta]$  of 0.01-5 dl/g as measured at 135°C in decalin and a softening temperature (TMA) of below 70°C, the weight ratio of (A)/(B) being from 100/0.1 to 100/10. There is also provided information recording base boards which have been molded from said cycloolefin type random copolymer compositions.

The cycloolefin type random copolymer compositions of the present invention and information recording base boards which are molded from said compositions are illustrated below in detail.

The cycloolefin type random copolymers [A] and [B] which constitute the compositions of the present invention are cycloolefin type random copolymers containing an ethylene component and a specific cycloolefin component.

In the cycloolefin type random copolymers, said cycloolefin component forms a structure represented by formula [III] or [IV]:



30 wherein n, m,  $\ell$  and  $R^1$  to  $R^{10}$  are as defined above.

The cycloolefin, i.e. a constituent component of the cycloolefin type random copolymers constituting the compositions of the present invention, is at least one cycloolefin selected from unsaturated monomers of formulae [I] and [II]. The cycloolefins of formula [I] can easily be prepared by condensation of cyclopentadienes with appropriate olefins by the Diels-Alder reaction, and similarly the cycloolefins of formula [II] can easily be prepared by condensation of cyclopentadienes with appropriate cycloolefins by the Diels-Alder reaction.

Examples of cycloolefins of formula [I] are given in Table 1 or, in addition to 1,4,5,8-dimethano-1,2,3,4,4a,5,8,8a-octahydronaphthalene, octahydronaphthalenes such as 2-methyl-1,4,5,8-dimethano-1,2,3,4,4a,5,8,8a-octahydronaphthalene, 2-ethyl-1,4,5,8-dimethano-1,2,3,4,4a,5,8,8a-octahydronaphthalene, 2-propyl-1,4,5,8-dimethano-1,2,3,4,4a,5,8,8a-octahydronaphthalene, 2-hexyl-1,4,5,8-dimethano-1,2,3,4,4a,5,8,8a-octahydronaphthalene, 2,3-dimethyl-1,4,5,8-dimethano-1,2,3,4,4a,5,8,8a-octahydronaphthalene, 2-methyl-3-ethyl-1,4,5,8-dimethano-1,2,3,4,4a,5,8,8a-octahydronaphthalene, 2-chloro-1,4,5,8-dimethano-1,2,3,4,4a,5,8,8a-octahydronaphthalene, 2-bromo-1,4,5,8-dimethano-1,2,3,4,4a,5,8,8a-octahydronaphthalene, 2-fluoro-1,4,5,8-dimethano-1,2,3,4,4a,8,8a-octahydronaphthalene, 2,3-dichloro-1,4,5,8-dimethano-1,2,3,4,4a,5,8,8a-octahydronaphthalene, 2-cyclohexyl-1,4,5,8-dimethano-1,2,3,4,4a,5,8,8a-octahydronaphthalene, 2-n-butyl-1,4,5,8-dimethano-1,2,3,4,4a,5,8,8a-octahydronaphthalene or 2-isobutyl-1,4,5,8-dimethano-1,2,3,4,4a,5,8,8a-octahydronaphthalene, and the compounds given in Table 2.

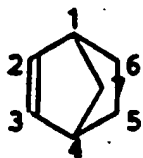
Table 1

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Chemical formula

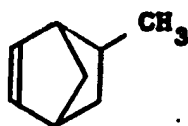
Compound name

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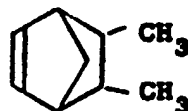
Bicyclo[2,2,1]hept-2-ene

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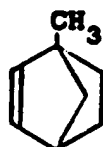
6-Methylbicyclo[2,2,1]hept-2-ene

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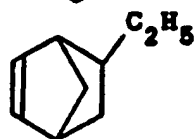
5,6-Dimethylbicyclo[2,2,1]hept-2-ene

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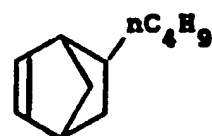
1-Methylbicyclo[2,2,1]hept-2-ene

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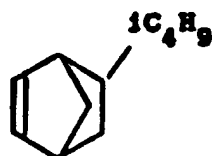
6-Ethylbicyclo[2,2,1]hept-2-ene

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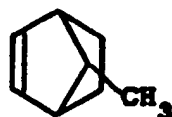
6-n-Butylbicyclo[2,2,1]hept-2-ene

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6-Isobutylbicyclo[2,2,1]hept-2-ene

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7-Methylbicyclo[2,2,1]hept-2-ene

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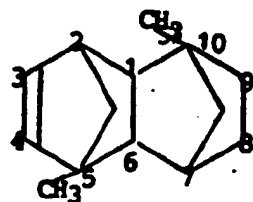
Table 2

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Chemical formula

Compound name

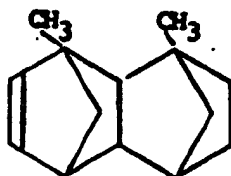
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5,10-Dimethyltetracyclo-  
[4.4.0.1<sup>2.5</sup>,1<sup>7.10</sup>]-3-dodecene

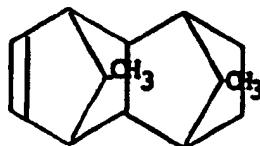
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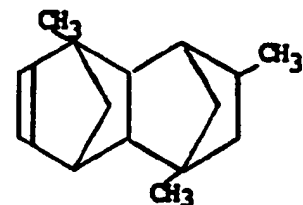
2,10-Dimethyltetracyclo-  
[4.4.0.1<sup>2.5</sup>,1<sup>7.10</sup>]-3-dodecene

25



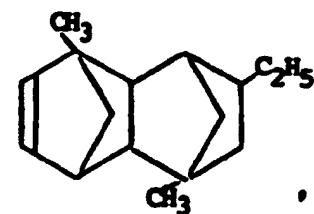
11,12-Dimethyltetracyclo-  
[4.4.0.1<sup>2.5</sup>,1<sup>7.10</sup>]-3-dodecene

30



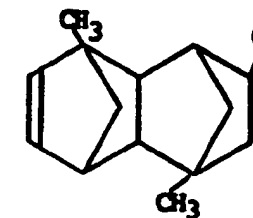
2,7,9-Trimethyltetracyclo-  
[4.4.0.1<sup>2.5</sup>,1<sup>7.10</sup>]-3-dodecene

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9-Ethyl-2,7-dimethyltetracyclo-  
[4.4.0.1<sup>2.5</sup>,1<sup>7.10</sup>]-3-dodecene

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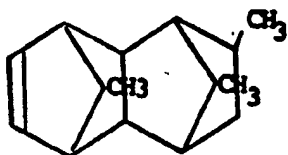
9-Isobutyl-2,7-dimethyltetracyclo-  
[4.4.0.1<sup>2.5</sup>,1<sup>7.10</sup>]-3-dodecene

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Table 2 (continued)

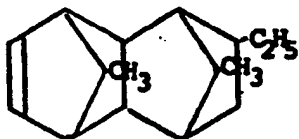
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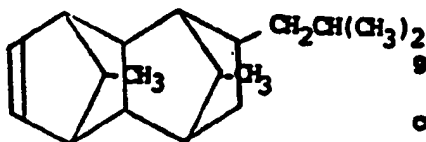
9,11,12-trimethyltetracyclo-  
[4,4,0,1<sup>2.5</sup>,1<sup>7.10</sup>]-3-dodecene

15



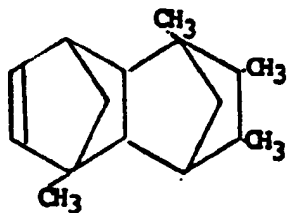
9-Ethyl-11,12-dimethyltetracyclo-  
[4,4,0,1<sup>2.5</sup>,1<sup>7.10</sup>]-3-dodecene

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9-Isobutyl-11,12-dimethyltetra-  
cyclo[4,4,0,1<sup>2.5</sup>,1<sup>7.10</sup>]-3-dodecene

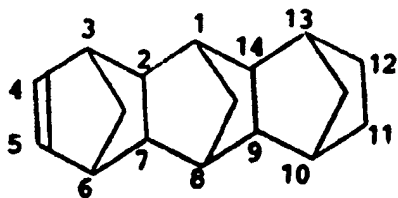
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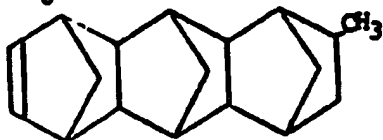
5,8,9,10-tetramethyltetracyclo-  
[4,4,0,1<sup>2.5</sup>,1<sup>7.10</sup>]-3-dodecene

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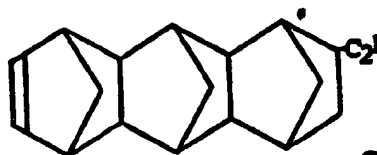
Hexacyclo[6,6,1,1<sup>3.6</sup>,1<sup>10.13</sup>,  
0<sup>2.7</sup>,0<sup>9.14</sup>]-4-heptadecene

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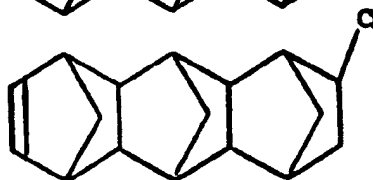
12-Methylhexacyclo[6,6,1,1<sup>3.6</sup>,  
1<sup>10.13</sup>,0<sup>2.7</sup>,0<sup>9.14</sup>]-4-heptadecene

45



12-Ethylhexacyclo[6,6,1,1<sup>3.6</sup>,  
1<sup>10.13</sup>,0<sup>2.7</sup>,0<sup>9.14</sup>]-4-heptadecene

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12-Isobutylhexacyclo[6,6,1,1<sup>3.6</sup>,  
1<sup>10.13</sup>,0<sup>2.7</sup>,0<sup>9.14</sup>]-4-heptadecene

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Table 2 (continued)

5		1,6,10-Trimethyl-12-isobutyl-
10		hexacyclo[6.8.1.1.3.6.1.10.13.0.2.7.0.9.14]-4-heptadecene
15		Octacyclo[8.8.0.1.2.9.1.4.7.1.11.18.1.13.16.0.3.8.0.12.17]-5-docosene
20		15-Methyloctacyclo[8.8.0.1.2.9.1.4.7.1.11.18.1.13.16.0.3.8.0.12.17]-5-docosene
25		
30		15-Ethyl-octacyclo[8.8.0.1.2.9.1.4.7.1.11.18.1.13.16.0.3.8.0.12.17]-5-docosene

Examples of cycloolefins of formula [II] are given in Tables 3 and 4.



Table 3

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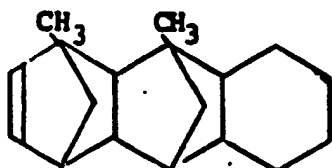
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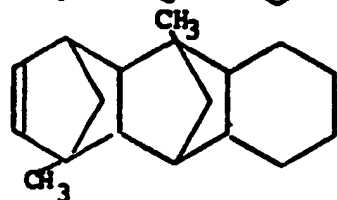
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## Chemical formula

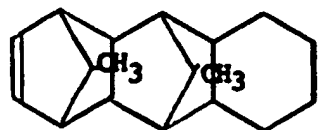
## Compound name



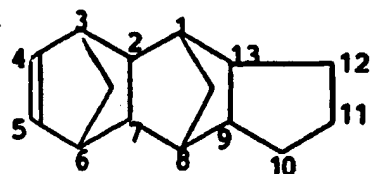
1,3-Dimethylpentacyclo[6,6,1,  
1<sup>3.6</sup>,0<sup>2.7</sup>,0<sup>9.14</sup>]-4-hexadecene



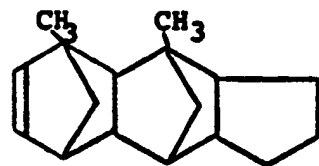
1,6-Dimethylpentacyclo[6,6,1,1<sup>3.6</sup>,  
0<sup>2.7</sup>,0<sup>9.14</sup>]-4-hexadecene



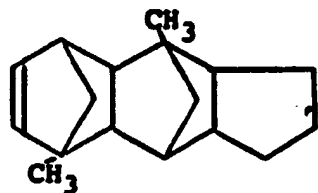
15,16-Dimethylpentacyclo[6,6,1,  
1<sup>3.6</sup>,0<sup>2.7</sup>,0<sup>9.14</sup>]-4-hexadecene



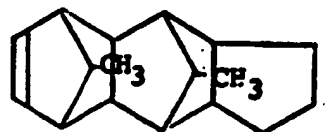
Pentacyclo[6,5,1,1<sup>3.6</sup>,0<sup>2.7</sup>,0<sup>9.13</sup>]-  
4-pentadecene



1,3-Dimethylpentacyclo[6,5,1,1<sup>3.6</sup>,  
0<sup>2.7</sup>,0<sup>9.13</sup>]-4-pentadecene



1,6-Dimethylpentacyclo[6,5,1,  
1<sup>3.6</sup>,0<sup>2.7</sup>,0<sup>9.13</sup>]-4-pentadecene

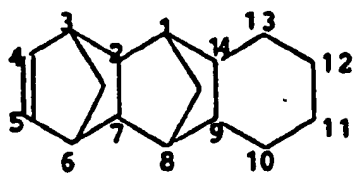


14,15-Dimethylpentacyclo[6,5,1,  
1<sup>3.6</sup>,0<sup>2.7</sup>,0<sup>9.13</sup>]-4-pentadecene

Table 3 (continued)

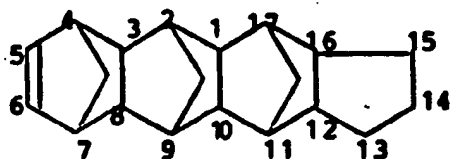
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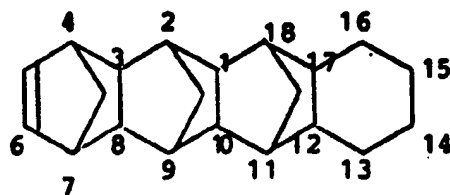
**Pentacyclo[6,6,1,1,3.6,0<sup>2.7</sup>,0<sup>9.14</sup>]-  
4-hexadecene**

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**Heptacyclo[8,7,0,1<sup>2.9</sup>,1<sup>4.7</sup>,1<sup>11.17</sup>,  
0<sup>3.8</sup>,0<sup>12.16</sup>]-5-eicosene**

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**Heptacyclo[8,8,0,1<sup>2.9</sup>,1<sup>4.7</sup>,1<sup>11.18</sup>,  
0<sup>3.8</sup>,0<sup>12.17</sup>]-5-heneicosene**

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Table 4

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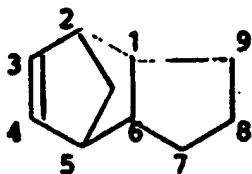
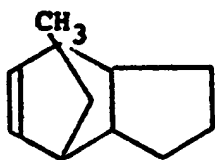
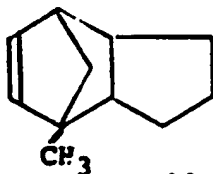
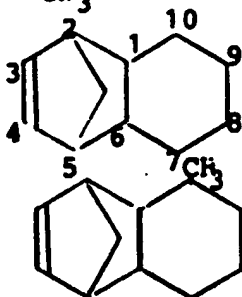
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## Chemical formula

## Compound name

Tricyclo[4,3,0,1<sup>2.5</sup>]-3-decene2-Methyl-tricyclo[4,3,0,1<sup>2.5</sup>]-  
3-decene5-Methyl-tricyclo[4,3,0,1<sup>2.5</sup>]-  
3-deceneTricyclo[4,4,0,1<sup>2.5</sup>]-3-undecene10-Methyl-tricyclo[4,4,0,1<sup>2.5</sup>]-  
3-undecene

The cycloolefin type random copolymers [A] and [B] which constitute the compositions of the present invention individually contain as essential components an ethylene component and the aforementioned cycloolefin component. In addition to the essential two components, however, the cycloolefin type random copolymers [A] and [B] may contain, if necessary, other copolymerizable unsaturated monomer components in such a range that they do not hinder the object of the present invention. Such unsaturated monomers which may optionally be copolymerized are  $\alpha$ -olefins of 3 to 20 carbon atoms, such as propylene, 1-butene, 4-methyl-1-pentene, 1-hexene, 1-octene, 1-decene, 1-dodecene, 1-tetradecene, 1-hexadecene, 1-octadecene or 1-eicosane, in the range of less than an equimolar amount of the ethylene component unit in the resulting random copolymer.

In the cycloolefin type random copolymer [A] which constitutes the compositions of the present invention, the repeating unit (a) derived from the ethylene component is preferably in the range of from 40 to 85 mol%, more preferably from 50 to 75 mol%, the repeating unit (b) derived from the cycloolefin component is preferably in the range from 15 to 60 mol%, more preferably from 25 to 50 mol%, and the repeating unit (a) derived from the ethylene component and the repeating unit (b) derived from the cycloolefin component generally form a substantially linear cycloolefin type random copolymer in which said repeating unit (a) and said repeating unit (b) are randomly orientated. When the cycloolefin type random copolymers of the present invention are substantially linear and do not have a gel-like cross-

linked structure, it can be confirmed by the fact that said copolymers perfectly dissolve in decalin kept at 135°C.

The intrinsic viscosity  $[\eta]$  as measured at 135°C in decalin of the cycloolefin type random copolymer [A] is 0.05-10 dl/g, preferably 0.08-5 dl/g, more preferably 0.2-3.0 dl/g, and particularly preferably 0.3-2.0 dl/g.

The softening temperature (TMA) as measured with a thermal mechanical analyzer of the cycloolefin type random copolymer [A] is not lower than 70°C, and is preferably 90-250°C, more preferably 100-200°C, particularly preferably 120-180°C, and still more particularly preferably 130-170°C. The glass transition temperature (T<sub>g</sub>) of said cycloolefin type random copolymer [A] is usually 50-230°C, preferably 70-210°C, more preferably 80-160°C, and particularly preferably 90-150°C.

The crystallinity index as measured by X-ray diffractometry of the cycloolefin type random copolymer [A] is preferably 0-10%, more preferably 0-7%, and most preferably 0-5%.

In the cycloolefin type random copolymer [B] which constitutes the compositions of the present invention, the repeating unit (a) derived from the ethylene component is preferably in the range of 60-98 mol%, more preferably 60-95 mol%, the repeating unit (b) derived from the cycloolefin component is preferably in the range of 2-40 mol%, more preferably 5-40 mol%, and the repeating unit (a) derived from the ethylene component and the repeating unit (b) derived from the cycloolefin component generally form a substantially linear cycloolefin type random copolymer in which said repeating unit (a) and said repeating unit (b) are random orientated. When the cycloolefin type random copolymers of the present invention are substantially linear and do not have a gel-like cross-linked structure, it can be confirmed by the fact that said copolymers perfectly dissolve in decalin kept at 135°C.

The intrinsic viscosity  $[\eta]$  as measured at 135°C in decalin of the cycloolefin type random copolymer [B] is 0.01-5 dl/g, preferably 0.08-3 dl/g, more preferably 0.2-3.0 dl/g, and particularly preferably 0.3-2.0 dl/g.

The softening temperature (TMA) as measured with a thermal mechanical analyzer of the cycloolefin type random copolymer [B] is below 70°C, and is preferably 60 to -10°C, more preferably 55 to -10°C. In this connection, the softening temperature of the cycloolefin type random copolymer [B] is suitably lower by 30-250°C, preferably 60-200°C, more preferably 80-190°C, and particularly preferably 90-160°C, than that of the cycloolefin type random copolymer [A]. The glass transition temperature (T<sub>g</sub>) of the cycloolefin type random copolymer [B] is generally -30 to 50°C, preferably -20 to 40°C. Furthermore, it is preferable that the glass transition temperature of the cycloolefin type random copolymer [B] is lower by 30-240°C, preferably 50-200°C, than that of the cycloolefin type random copolymer [A].

The crystallinity index as measured by X-ray refractometry of the cycloolefin type random copolymer [B] is preferably 0-10%, more preferably 0-7%, and most preferably 0-5%.

In the compositions, the weight ratio of the cycloolefin type random copolymer [A] / cycloolefin type random copolymer [B] is 100/0.1 to 100/10, preferably 100/0.3 to 100/7, more preferably 100/0.5 to 100/5, and still particularly preferably 100/0.7 to 100/4. The present compositions have insufficient precision moldability as well as an insufficient improvement in adhesion between the molded articles of said compositions and information recording films to be formed thereon when said weight ratio is less than 100/0.1, and said molded articles have a poor transparency or a lowered surface smoothness when said weight ratio is greater than 100/10, thus decreasing the performance of the compositions as information recording materials.

The cycloolefin type random copolymers [A] and [B] may both be prepared by suitably selecting the conditions under which they are prepared in accordance with the processes as proposed by the present applicant in Japanese Patent L-O-P Publns. Nos.168708/1985, 120816/1986, 115912/1986, 115916/1986, 95905/1986, 95906/1986, 271308/1986 and 272216/1986.

Various known processes may be used to prepare the cycloolefin type random copolymer compositions of the present invention, for example, a process wherein the cycloolefin type random copolymers [A] and [B] are prepared separately, and the copolymers [A] and [B] thus prepared are blended by means of an extruder to obtain a desired composition, a solution blending process wherein the copolymers [A] and [B] are thoroughly dissolved separately in suitable solvent, for example, saturated hydrocarbons such as heptane, hexane, decane or cyclohexane, or aromatic hydrocarbons such as toluene, benzene or xylene, and the respective solutions are subjected to solution blending to obtain a desired composition, or a process wherein the copolymers [A] and [B] are prepared individually by means of separate polymerization reactors, and the resulting polymers are blended in a separate vessel to obtain a desired composition.

The intrinsic viscosity  $[\eta]$  as measured at 135°C in decalin of the cycloolefin type random copolymer compositions of the present invention is 0.05-10 dl/g, preferably 0.08-5 dl/g, more preferably 0.2-3 dl/g, and still particularly preferably 0.3-2 dl/g, the softening temperature (TMA) as measured with a thermal mechanical analyzer of said compositions is 80-250°C, preferably 100-200°C, and more preferably 120-170°C, and the glass transition temperature (T<sub>g</sub>) of said compositions is 70-230°C, preferably 80-160°C, and more preferably 90-150°C.

The cycloolefin type random copolymer compositions of the present invention contain as essential components the aforesaid cycloolefin type random copolymer [A] and cycloolefin type random copolymer [B]. In addition to the above-mentioned essential two components, however, the present compositions may incorporate, for example heat

stabilizers, weathering stabilizers, antistatic agents, slip agents, anti-blocking agents, anti-fogging agents, lubricants, dyes, pigments, natural oil, synthetic oil, or a wax. The amounts of these additives may be suitably decided. For instance, the stabilizers which may be optionally incorporated include phenolic antioxidants such as tetrakis [methylene-3-(3,5-di-*t*-butyl-4-hydroxyphenyl)propionate] methane,  $\beta$ -(3,5-di-*t*-butyl-4-hydroxyphenyl)propionic acid alkyl ester (particularly preferred are alkyl esters of less than 18 carbon atoms) or 2,2'-oxamidobis[ethyl-3-(3,5-di-*t*-butyl-4-hydroxyphenyl)propionate], metallic salts of fatty acid such as zinc stearate, calcium stearate or calcium 12-hydroxystearate, and fatty esters of polyhydric alcohols such as glycerin monostearate, glycerin monolaurate, glycerin distearate, pentaerythritol distearate or pentaerythritol tristearate. These compounds may be incorporated into the present compositions either singly or in combination. For instance, there may be used a combination of tetrakis[methylene-3-(3,5-di-*t*-butyl-4-hydroxyphenyl)propionate]methane with zinc stearate and glycerin monostearate.

In the present invention, it is preferable to use phenolic antioxidants in combination with fatty esters of polyhydric alcohols. The fatty esters of polyhydric alcohols are preferably those which are obtained by esterification of a part of the alcoholic hydroxyl groups of polyhydric alcohols having a valence of at least 3. Examples of such fatty esters of polyhydric alcohols include fatty esters of glycerin such as glycerin monostearate, glycerin monolaurate, glycerin monomyristate, glycerin monopalmitate, glycerin distearate or glycerin dilaurate, and their mixtures, fatty esters of pentaerythritol such as pentaerythritol monostearate, pentaerythritol monolaurate, pentaerythritol distearate, pentaerythritol dilaurate or pentaerythritol tristearate, and their mixtures, and fatty esters of sorbitol such as sorbitol monostearate, sorbitol monolaurate or sorbitol distearate, and their mixture.

The phenolic antioxidants as illustrated above are generally used in an amount of 0.01-10 parts by weight, preferably 0.05-3 parts by weight based on 100 parts by weight of the cycloolefin type random copolymer composition. Similarly, the fatty esters of polyhydric alcohols are generally used in an amount of 0.01-10 parts by weight, preferably 0.05-3 parts by weight based on 100 parts by weight of the said composition.

The stabilizers which comprise a combination of the above-mentioned phenolic antioxidant and fatty ester of a polyhydric alcohol, when added to the composition containing the aforementioned cycloolefin type random copolymers [A] and [B], not only can impart excellent stability to said composition but also can impart excellent stability to said copolymer [A] and also to said copolymer [B] even when said stabilizers are added individually to said copolymers [A] and [B].

In this connection, stabilizers comprising a combination of the aforementioned phenolic antioxidant and fatty ester of a polyhydric alcohol as mentioned above can also impart excellent stability to a polymer, such as polymethyl methacrylate (PMMA), polycarbonate (PC) or poly-4-methylpentene-1, when said stabilizers are added thereto.

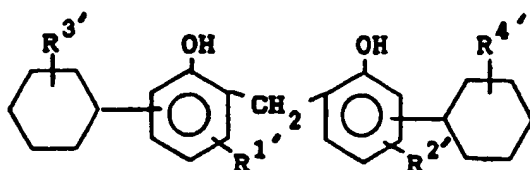
In order to prepare a thin optical disc having good appearance and a broad projection area by injection molding of a cycloolefin random copolymer [A], a cycloolefin random copolymer [B], polymethyl methacrylate, polycarbonate or poly-4-methylpentene-1, it is preferred to incorporate therein at least one stabilizer selected from:

- (1) phenolic antioxidants having in the molecule a beta-(3,5-di-*tert*-butyl-4-hydroxyphenyl) propionyl group;
- (2) normally liquid hydrocarbons having a molecular weight of about 200 to 3000;
- (3) phenolic antioxidants having a cycloalkyl group; and
- (4) esters of bis(dialkylphenyl) pentaerythritol diphosphite.

Examples of the phenolic antioxidants (1) include tetrakis [methylene(3,5-di-*tert*-butyl-4-hydroxyphenyl) propionate] methane, alkyl beta-(3,5-di-*tert*-butyl-4-hydroxyphenyl) propionates (as the alkyl, those having up to 18 carbon atoms, particularly methyl, are preferred) and 2,2'-oxamidobis [ethyl-3-(3,5-di-*tert*-butyl-4-hydroxyphenyl)propionate].

The hydrocarbons (2) include naturally occurring and synthesized aliphatic, alicyclic, aromatic and partly hydrogenated aromatic hydrocarbons, optionally substituted by alkyl, aryl and/or aralkyl groups. Examples of such hydrocarbons include dodecylbenzene, polyalkylbenzenes by-produced in the preparation of a dodecylbenzene oil, dibenzylbenzenes, alkyl-naphthalenes, polyphenylalkane oil, alkylated tetralines, olefin oligomers such as ethylene-, propylene-, ethylene-propylene- and butene-oligomers, fluid paraffin, squalane and mineral oils.

The cycloalkyl-containing phenolic antioxidants (3) include a compound of formula:



wherein R<sup>1'</sup>, R<sup>2'</sup>, R<sup>3'</sup> and R<sup>4'</sup> are the same or different and each represents hydrogen or alkyl. Commercially available

cycloalkyl-containing antioxidants which can be used as a stabilizer include 2,2'-dihydroxy-3,3'-dicyclohexyl-5,5'-dimethyl-diphenylmethane (Vulkanox ZKF, supplied by Farbenfabriken Bayer AG), 2,2'-hydroxy-3,3'-di-(alpha-methylcyclohexyl)-5,5'-dimethyl-diphenylmethane (Nomox WSP, supplied by Seiko Kagaku KK), 1,1-bis(4-hydroxyphenyl) cyclohexane (Nomox WSL, supplied by Seiko Kagaku KK) and 2-(1-methylcyclohexyl)-4,6-dimethylphenol (Antigenew, supplied by Sumitomo Kagaku KK).

The stabilizers illustrated above may be used alone or in combination. The stabilizer is normally used in an amount of from 0.01 to 5 parts, preferably from 0.05 to 3 parts, and most preferably from 0.1 to 1 parts by weight per 100 parts by weight of the polymer.

Information recording base boards in accordance with the present invention are prepared by molding the cycloolefin type random copolymer compositions into desired forms in the usual way, and laminating according to a conventional method information recording films on the thus molded information recording bases.

The information recording base boards of the present invention may have all sorts of structures, for example, they may be in the form of optical disc, flexible optical discs, optical cards, optical fibers, optical tapes, photoconductive halos or magnetic discs.

Conventionally known information recording films can be laminated on the information recording bases of the present invention. Examples of useful information recording films include tellurium type alloys such as TeSe, TeSePb, Se, TcSeSb or TeGeSb, films having dispersed metals in organic matrixes such as Te-carbon, gold alloys such as AuPd or AuPt, terbium type alloys such as TbFe, TbFeCo or TbGdFe, cyanine or naphthoquinone type organic colour films, and magnetic materials such as ferrite. As refractive materials used in such information recording films, there may be mentioned metals such as Al, Cr, Au or Co, and alloys thereof. Examples of protective or enhance films for such information recording films are those which comprise  $\text{Si}_3\text{N}_4$ ,  $\text{SiO}_2$ , ZnO, TiO, ZnSe or CdS. In laminating such information recording films on the information recording bases of the present invention, there may be employed any conventionally known procedures, for example vacuum deposition, sputtering, ion plating, spin coating or coater coating.

The cycloolefin random copolymer compositions according to the invention have applications in various fields, including optical lenses, transparent windows, flenel lenses, array lenses for a laser, optical fiber connectors, recording sheets for photography, films, plane lenses of distributed refractive indexes (grated lenses), optical materials of reformed surface hardness (materials having dispersed therein glass or ceramics of the same refractive index), polarized light filters, filters for selection of wavelength, photo sensitive films, covering and capping agents for LED and semiconductor lasers, mirrors, prisms, beam splitters, EL bases and other optical applications, water tanks of electric irons, parts of electronic ranges, base plates for liquid crystal display instruments, base plates of flexible prints, base plates for high frequency circuits, electro-conductive sheets and films, film condensers, insulating painted films, materials for lightening instruments, window materials for display elements, housings of electrical instruments, protective films for lithography and other electrical applications, syringes, pipettes, animal cages, body thermometers, beakers, laboratory dishes, measuring cylinders, bottles, artificial joints, carriers for use in chromatography and other chemical and medical applications, membranes for gas separation, membranes for ultrafiltration, membranes for reverse osmosis, membranes for gas-liquid separation and other membranes for separation purpose, bodies of cameras, housings of various metering instruments, films, helmets, toys and stationery. They can also be used in applications where shape-remembering properties are required and as vibration dampers and tubes, including for example, joints for pipes of different shapes, lamination materials to be applied inside or outside of pipes or rods, optical fiber connectors, fastening pins, gypsums, vessels, automotive bumpers, various gap fillers, vibration dampers (sound insulators) for coating metallic surfaces, medical tubes, wrapping films, protective films (for metallic plates and pipes) and heat-sealable films. They are also useful as candles, impregnating agents for matches, paper finishing agents, sizes, antioxidants for rubber, water-proofing agent for cardboards, slow releasing agents for chemical fertilizers, heat accumulators, binders for ceramics, paper condensers, electric insulating materials for electric wires and cables, agents for decelerating neutrons, fiber finishing aids, water-repellents for building materials, protecting agents for coatings, polishing agents, a thixotropy imparting agents, core hardening agents for pencils and crayons, carbon ink substrates, electrophotographic toners, lubricants and releasing agents for molding of synthetic resins, resin coloring agents, hot-melt adhesives, and lubricant greases.

The cycloolefin type random copolymer compositions of the present invention have excellent transparency, heat resistance, heat ageing characteristics, solvent resistance, dielectric characteristics and mechanical characteristics as well as precision moldability, and the information recording bases molded from the present cycloolefin type random copolymer compositions have excellent adhesion to information recording films to be laminated on said information recording bases.

The present invention is illustrated below in more detail in the Examples. Physical properties indicated in the Examples were measured or evaluated according to the following procedures:

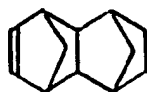
(1) Softening temperature (TMA): Using a Thermo Mechanical Analyzer (manufactured and sold by Du Pont), the

softening temperature was measured in terms of heat deformation behavior of a test sheet 1 mm thick. A quartz needle placed vertically on the test sheet had applied to it a load of 49 g, while elevating the temperature of the test sheet. The temperature at which the needle penetrated 0.635 mm into the test sheet was taken to be the TMA. (2) Adhesion of information recording film: Using a thermohydrostat (PI-3G, manufactured and sold by Tabai Co.), a resin base having an information recording film attached thereto was treated for one week at 85°C and 85% RH. Immediately after the treatment, the surface profile of the recording film and adhesion between the recording film and resin base were observed under a metallurgical microscope (magnifying power 100-400 times).

#### Polymerization Example 1

Preparation of copolymer (A) having a softening temperature of at least 70°C

With a 2-litre glass polymerization reactor equipped with a stirring blade, there was carried out continuously a copolymerization between ethylene and 1,4,5,8-dimethano-1,2,3,4,4a,5,8,8a-octahydronaphthalene (structural formula:



hereinafter abbreviated to DMON). That is, into the polymerization reactor were continuously charged a cyclohexane solution of DMON so that the DMON concentration of the solution in the polymerization reactor was 60 g/l, a cyclohexane solution of VO(OC<sub>2</sub>H<sub>5</sub>)Cl<sub>2</sub> as a catalyst so that the vanadium concentration of the solution in the polymerization reactor was 0.9 mmol/l, and a cyclohexane solution of ethylaluminum sesquichloride (Al(C<sub>2</sub>H<sub>5</sub>)<sub>1.5</sub>Cl<sub>1.5</sub>) so that the aluminium concentration of the solution in the polymerization reactor was 7.2 mmol/l, while continuously withdrawing from the bottom of the polymerization reactor the polymerization liquid so that the volume of the polymerization liquid in the polymerization reactor was constantly 1 litre. Simultaneously, into the polymerization reactor from the top thereof, ethylene was fed at a rate of 85 l/hr, hydrogen was fed at a rate of 6 l/hr and nitrogen was fed at a rate of 45 l/hr. The copolymerization was carried out at 10°C by circulating a refrigerant through a jacket fitted externally to the polymerization reactor.

A polymerization reaction mixture containing an ethylene DMON random copolymer was obtained. The polymerization reaction was stopped by adding a small amount of isopropyl alcohol to the polymerization liquid to be withdrawn. Thereafter, an aqueous solution composed of 1 liter of water and 5 ml of concentrated hydrochloric acid and the polymer solution were contacted at a ratio of 1:1 with strong stirring by a homomixer to transfer the catalyst residue to the aqueous layer. The mixture was left to stand, and the water layer was removed. The residue was washed twice with distilled water to purify and separate the polymer solution. Thereafter, the polymerization liquid was poured into a household mixer containing acetone in about three times the volume of the polymerization liquid, while rotating the mixer, thereby depositing the resulting copolymer. The deposited copolymer was collected by filtration, dispersed in acetone so that the polymer concentration was about 50 g/l, and the copolymer was treated at the boiling point of acetone. After the treatment the copolymer was collected by filtration and dried at 120°C overnight under reduced pressure.

The thus obtained ethylene DMON-random copolymer (A) had an ethylene unit of 59 mol% as measured by <sup>13</sup>C-NMR analysis, an intrinsic viscosity [η] of 0.42 dl/g as measured at 135°C in decalin, and a softening temperature (TMA) of 154°C.

#### Polymerization Example 2

Preparation of copolymer (B) having a softening temperature of below 70°C

The same copolymerization as in Polymerization Example 1 was carried out except that the cyclohexane solutions containing DMON, VO(OC<sub>2</sub>H<sub>5</sub>)Cl<sub>2</sub> and ethylaluminum sesquichloride, respectively, were fed into the polymerization reactor so that the concentrations of DMON, VO(OC<sub>2</sub>H<sub>5</sub>)Cl<sub>2</sub> and ethylaluminum sesquichloride in said solutions in the polymerization reactor were 23 g/l, 0.7 mmol/l and 5.6 mmol/l respectively and in that the ethylene, hydrogen and nitrogen were fed into the polymerization reactor at rates of 140 l/hr, 13 l/hr and 25 l/hr, respectively. After the completion of the copolymerization, the resulting copolymer was deposited and collected in the same manner as in Polymerization Example 1, and then dried for 12 hours at 180°C under reduced pressure.

The thus obtained ethylene DMON-random copolymer (B) had an ethylene unit of 89 mmol% as measured by  $^{13}\text{C}$ -NMR analysis, an intrinsic viscosity  $[\eta]$  of 0.44 dl/g as measured at 135°C in decalin, and a softening temperature (TMA) of 39°C.

#### Experiment Run No. 1

400 g of the copolymer (A) and 4 g of the copolymer (B) (weight ratio: (A)/(B) = 100/1) were poured into 8 litres of cyclohexane and dissolved at about 50°C with thorough stirring to obtain a homogeneous solution. The thus obtained homogeneous solution was poured in 24 litres of acetone to deposit an (A)/(B) blend. The thus obtained blend was dried at 120°C under reduced pressure overnight.

The (A)/(B) blend thus obtained was incorporated with 0.5%, 0.05% and 0.5%, based on the total weight of the resins [A] and [B], of tetrakis[methylene-3-(3,5-di-*t*-butyl-4-hydroxyphenyl)propionate] methane, zinc stearate and glycerin monostearate, respectively, as stabilizers, and then pelletized at 23°C with a 20 mm $\phi$  extruder (L/D=20). Thereafter, the pellets thus obtained were molded with an injection molding machine IS-50 manufactured and sold by Toshiba Machine Co., Ltd. into a disc of 1 mm thickness and 80 mm $\phi$  (both surfaces thereof were mirror finished). The disc thus obtained was then subjected to ultrasonic cleaning with Freon TE (a product of Mitsui Du Pont Fluorochemical Co.), and then a three-layered information recording film was provided on the surface by a sputtering method with  $\text{Si}_3\text{N}_4/\text{TbFeCo}/\text{Si}_3\text{N}_4$  (without pretreatment such as sputtering; the film thickness was 500 Å including a core layer). The disc obtained was allowed to stand at 85°C and 85% RH for 170 hours in a thermo-hydrostat. Microscopic observation of the surface state of the recording film revealed that no change in appearance of the recording film was observed in comparison with the appearance of said film prior to the test and the adhesion of the recording film to the disc was found to be favorable.

#### Experiment Run No. 2

In the same manner as in Experiment Run No. 1, the copolymer (a) obtained in Polymerization Experiment Run No. 1 was incorporated with the stabilizers, pelletized, and formed into a disc and the information recording film was then provided on the surface. Thereafter, the disc thus obtained was evaluated in the same way as in Experiment Run No. 1. As a result of microscopic observation of the recording film surface after the environmental test, there were observed a portion in the vicinity of the disc surface where foreign matter was present and the peel of the recording film at a portion of the disc surface where Freon liquid remains (Freon liquid sagging portion) formed at the time of cleaning the disc.

#### Experiment Run No. 3

Experiment Run No. 2 was repeated except that the disc obtained was allowed to stand at 60°C, and 85% RH for 170 hours before evaluation.

#### Experiment Run No. 4

A blend was prepared by repeating Experiment Run No. 1 except that the (A)/(B) weight ratio employed was changed to 100/15. The evaluation of the information recording disc obtained was conducted in the same way as in Experiment Run No. 1. As the result, the state of the film before and after the environmental test was found to be favorable. However, the disc had poor transparency and use as an information recording disc was found to be difficult.

#### Experiment Runs Nos. 5-10

Information recording discs were prepared by using copolymer (A) and (B) which were synthesized using the monomers shown in Table 5 in the same manner as Polymerization Experiment Run Nos. 1 and 2 and the evaluation of the resulting information recording discs were conducted in the same way as in Experiment Run No. 1. The results are shown in Table 5.

#### Experiment Runs Nos. 11-13

Information recording discs were prepared by using the 80 mm $\phi$  discs (after cleaning with Freon ET) respectively prepared in Experiment Runs Nos. 1, 6 and 8, which were individually provided as an information recording film with a  $\text{SiO}_2$  single layer film (film thickness: 500 Å) on their surface in place of the  $\text{Si}_3\text{N}_4/\text{TbFeCo}/\text{Si}_3\text{N}_4$  three-layered film. After the same environmental test as in Experiment Run No. 1, the surface of the recording film of each disc was



## EP 0 291 208 B1

observed with a microscope. It was found that the adhesion of the film to the disc was favorable.

### Experiment Runs Nos. 14-15

5        On the surface of 80 mm $\phi$  discs respectively prepared in Experiment Runs Nos. 6 and 9, Al was deposited by a vacuum deposition method to form information recording films (thickness: 100 Å). After subjecting the information recording discs to the same environmental test as in Experiment Run No. 1, the surface of the recording film of each disc was observed with a microscope. It was found that the adhesion between the deposited Al and disc was good.

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Table 8











Ex. Run No.	Copolymer (A)				Copolymer (B)			
	Cycloolefin	Third monomer	Composition (mole%)		Cycloolefin	Third monomer	Composition (mole%)	
			Styrene	Third monomer			Styrene	Third monomer
1		—	89	—		—	89	—
5	•	—	•	—	•	—	•	—
6	•	—	•	—	•	—	•	—
7	•	—	•	—		—	72	—
8		Butene	89	0.8		—	89	—
9	•	•	•	•	•	Butene	74	1.2
10		—	82	—		—	89	—
11		—	89	—	•	—	•	—
12	•	—	•	—	•	—	•	—
13	•	Butene	89	0.9	•	—	•	—
14	•	—	89	—	•	—	•	—
15	•	Butene	89	0.9	•	Butene	74	1.2
2 and 3		—	89	—	—	—	—	—
4	•	—	•	—		—	89	—

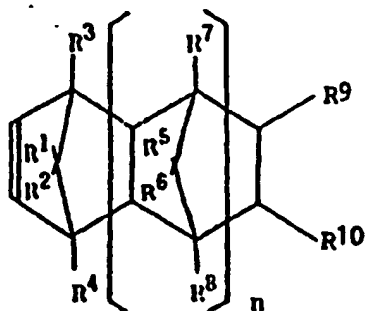
Table 5 (Continued)

Ex. Run No.	Blending ratio (A)/(B) (weight ratio)	Transparency of 80mm $\phi$ disc	State of film after environmental test
1	100/1	Good	Good adhesion
5	100/0.5	"	"
6	100/2	"	"
7	100/1.5	"	"
8	100/1	"	"
9	100/3	"	"
10	100/1.5	"	"
11	100/1	"	"
12	100/2	"	"
13	100/1	"	"
14	100/2	"	"
15	100/3	"	"
2	100/0	Good	Peeling of recording film occurs at Freon liquid sagging portion and/or portion where foreign matters exist.
3	100/0	Good	Good adhesion
4	100/15	Bad	Good adhesion

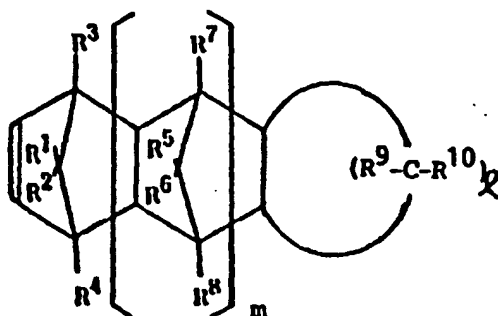
## Claims

## 1. A composition comprising

(A) a random copolymer containing units derived from ethylene and units derived from a cycloolefin of formula [I] or [II],



[I]



[II]

wherein  $n$  and  $m$  are each 0 or a positive integer,  $l$  is an integer of at least 3, and  $R^1$  to  $R^{10}$  each represent hydrogen, halogen or a hydrocarbon group, said random copolymer having an intrinsic viscosity  $[\eta]$  of 0.05-10 dl/g as measured at 135°C in decalin and a softening temperature (TMA) not lower than 70°C, and  
(B) a random copolymer containing units derived from ethylene and units derived from a cycloolefin of said formula [I] or [II] and having an intrinsic viscosity  $[\eta]$  of 0.01-5 dl/g as measured at 135°C in decalin and a softening temperature (TMA) of below 70°C, the weight ratio of (A)/(B) being from 100/0.1 to 100/10.

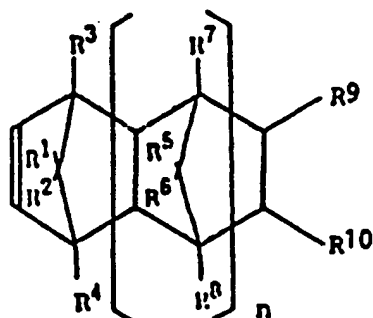
2. A composition according to claim 1 which also comprises a phenolic antioxidant and a fatty ester of a polyhydric alcohol as stabilizers.
3. A composition according to claim 2 wherein the fatty ester of a polyhydric alcohol has been obtained by the partial esterification of a polyhydric alcohol having at least three hydroxyl groups.
4. An information recording base board which has been molded from a composition as claimed in any one of the preceding claims.

## Patentansprüche

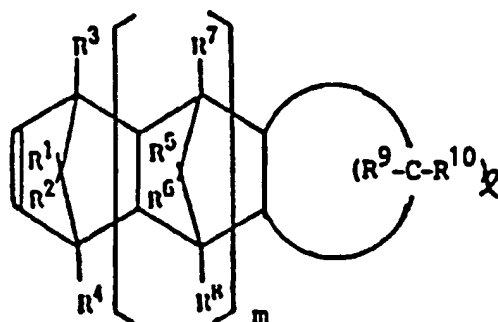
## 1. Masse, umfassend

(A) ein statistisches Copolymer, enthaltend Einheiten, die abgeleitet sind von Ethylen und Einheiten, die ab-

geleitet sind von einem Cycloolefin der Formel [I] oder [II]



[I]



[II]

wobei  $n$  und  $m$  jeweils 0 oder eine positive ganze Zahl sind,  $\ell$  eine ganze Zahl von mindestens 3 ist und  $R^1$  bis  $R^{10}$  jeweils Wasserstoff, Halogen oder eine Kohlenwasserstoffgruppe bedeuten, wobei das statistische Copolymer eine Grundviskosität  $[\eta]$  von 0,05-10 dl/g, gemessen bei 135°C in Decalin, und eine Erweichungstemperatur (TMA) nicht unter 70°C aufweist, und

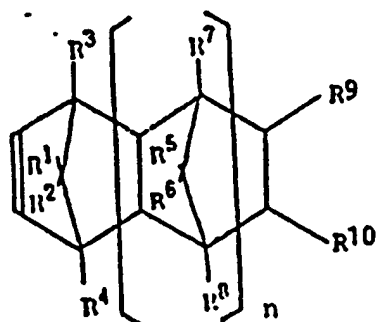
(B) ein statistisches Copolymer, enthaltend Einheiten, die abgeleitet sind von Ethylen und Einheiten, die abgeleitet sind von einem Cycloolefin der Formel [I] oder [II] mit einer Grundviskosität  $[\eta]$  von 0,01-5 dl/g, gemessen bei 135°C in Decalin, und einer Erweichungstemperatur (TMA) unter 70°C, wobei das Gewichtsverhältnis (A)/(B) 100/0,1 bis 100/10 beträgt.

2. Masse nach Anspruch 1, die zusätzlich ein phenolisches Antioxidans und einen Fettsäureester eines mehrwertigen Alkohols als Stabilisatoren enthält.
3. Masse nach Anspruch 2, wobei der Fettsäureester eines mehrwertigen Alkohols erhalten worden ist durch teilweise Veresterung eines mehrwertigen Alkohols mit mindestens drei Hydroxylgruppen.
4. Informationsaufzeichnungs-Grundplatte, die geformt worden ist aus einer Masse nach einem der vorangehenden Ansprüche.

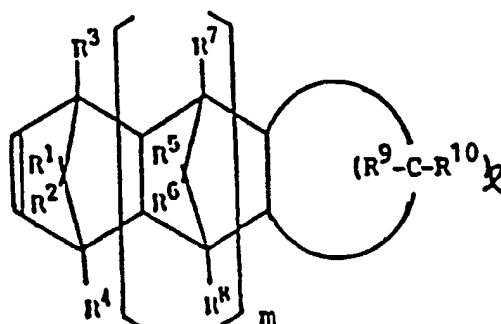
#### Revendications

1. Composition comprenant :

(A) un copolymère statistique contenant des motifs dérivés de l'éthylène et des motifs dérivés d'une cyclooléfine qui répond aux formules [I] et [II] :



[I]



[II]

dans lesquelles n et m représentent chacun 0 ou un nombre entier positif, représente un nombre entier valant au moins 3, et les symboles R<sup>1</sup> à R<sup>10</sup> représentent chacun un atome d'hydrogène, un atome d'halogène ou un groupe hydrocarboné, ledit copolymère statistique présentant une viscosité intrinsèque  $[\eta]$  comprise entre 0,05 et 10 dl/g, telle que mesurée à 135° C dans la décane, et une température de ramollissement (TMA) qui n'est pas inférieure à 70° C, et

(B) un copolymère statistique contenant des motifs dérivés de l'éthylène et de motifs dérivés d'une cyclo-oléfine qui répond à ladite formule [I] ou [II], et présentant une viscosité intrinsèque  $[\eta]$  comprise entre 0,01 et 5 dl/g, telle que mesurée à 135° C dans la décane, et une température de ramollissement (TMA) inférieure à 70° C, le rapport pondéral de (A)/(B) étant compris entre 100/0,1 et 100/10.

2. Composition conforme à la revendication 1, qui comprend également un antioxydant phénolique et un ester d'acide gras et d'un polyalcool comme stabilisants.
3. Composition conforme à la revendication 2, dans laquelle on a obtenu l'ester d'acide gras et d'un polyalcool par estérification partielle d'un polyalcool présentant au moins trois groupes hydroxyle.
4. Support d'enregistrement d'information qui a été moulé à partir d'une composition conforme à l'une quelconque des précédentes revendications.